

# FROM THE **CHIEF SCIENTIST'S DESK**

**J. Robert Schrieffer**



In this issue, path-breaking studies of the so-called FFLO state of superconductors are reported for the first time. In conventional superconductors, electrons of opposite spin and momentum are paired to produce the superconducting condensate. In the FFLO state the electron spin Zeeman energy favors pairing parallel spins to some extent. The orbital state of the superconductor develops nodes in real space leading to alternating layers of superconducting material and spin-polarized magnetic walls. Subsequently, Buzdin and Brison extended the FFLO theory to include interaction of orbital and paramagnetic effects. One finds that in addition to spatially modulated structures the superconducting gap also takes on higher Landau level structure with finite angular momentum. In this case a sequence of first order transitions takes place within the FFLO phase as the external magnetic field is increased. These transitions should be observed in the magnetization or critical current as jumps that separate states of different angular momenta.

These studies were carried out on samples of  $\text{CeCoIn}_5$  grown by John Sarrao at LANL. This material has a layered structure that inhibits electronic orbital motion for magnetic field parallel to the conducting planes. The large spin susceptibility of  $\text{CeCoIn}_5$  favors the paramagnetic effects of the FFLO mechanism.

These techniques were used to study the effects, namely heat capacity, magnetization, and penetration depth. The first method gives a direct measure of the thermodynamic state of the material, in particular, the phase transitions. The second method employs a torque method using a cantilever that acts as one plate of a capacitor. The third technique uses a contactless cavity technique that alters its resonant frequency as a function of the state of the superconductor. This is directly proportional to the penetration depth of the magnetic field.

These measurements constitute the first measurements of the FFLO state and demonstrate a fundamentally new state of a superconductor in a strong magnetic field, a long sought goal.

## **Fulde-Ferrell-Larkin-Ovchinnikov Superconductivity in $\text{CeCoIn}_5$**

H.A. Radovan, S.W. Tozer, T.P. Murphy, S.T. Hannahs, E.C. Palm, D. Hall, NHMFL

N.A. Fortune, Smith College, Physics Department

C. Martin and C.C. Agosta, Clark University, Physics Department

### **Historical Background**

Superconductivity, a macroscopic quantum phenomenon where electrons of opposite spin and momentum condense into pairs, was discovered in 1911 by Onnes, but eluded theoretical explanation until the work of Bardeen, Cooper, and Schrieffer in 1957. A sufficiently large magnetic field will destroy superconductivity by coupling to the orbital motion of the electrons. This orbital limit

(critical field) separates the uniform superconducting state from the normal metallic state. A magnetic field can, however, couple predominantly to the spins of the electrons. The superconductor is then in the paramagnetic limit. It was shown in 1964 by Fulde & Ferrell,<sup>1</sup> and independently, Larkin & Ovchinnikov<sup>2</sup> (FFLO), that this superconducting state would be fundamentally different from the conventional BCS case. In this new state, the magnetic field tries to polarize the opposite spins of the superconducting pairs. In response the superconducting order parameter develops nodes in real space, leading to alternating regions of superconducting layers and spin-polarized magnetic walls. This FFLO state manifests itself as a wedge in the field-temperature (B-T) phase diagram at very low temperatures just below the critical field. The exact shape of the B-T phase diagram depends on many microscopic parameters, such as Fermi surface geometry, dimensionality of the host crystal, impurities, and ratio of orbital-to-paramagnetic effects.

In 1996, Buzdin and Brison<sup>3</sup> extended the FFLO theory to include the interaction of orbital and paramagnetic effects. They showed that in addition to becoming spatially modulated, the superconducting order parameter would also assume higher Landau level states with a finite angular momentum. Under these conditions, a cascade of first order phase transitions within the FFLO phase occurs with increasing external magnetic fields. Each transition should be observable in magnetization or critical current as a discontinuous step that separates sub-phases with different orbital quantum number.

The material in which we observed the FFLO state is the heavy fermion superconductor CeCoIn<sub>5</sub>, grown by John Sarrao at LANL.<sup>4</sup> CeCoIn<sub>5</sub> has a critical temperature of 2.3 K and a layered electronic structure. This anisotropy inhibits orbital motion of the electrons for fields aligned exactly parallel to the conducting planes. Additionally, the large spin susceptibility favors paramagnetic limitation. Thus, this material fulfills the delicate balance of properties needed to observe FFLO superconductivity.

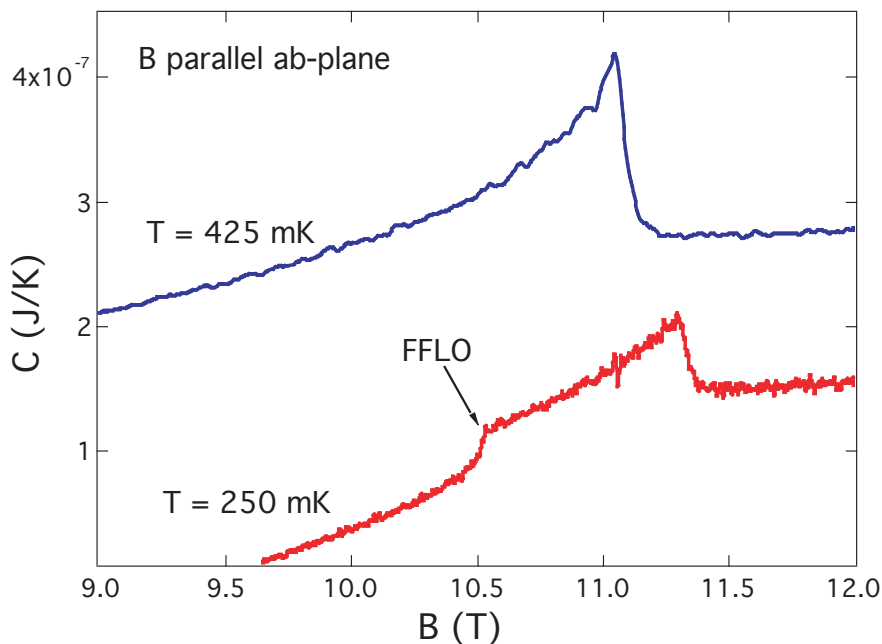
## Experimental Methods

Two of the three techniques used to perform the heat capacity, magnetization, and penetration depth studies presented here were developed with funding from the Visiting Scientist Program and the In-House Research Program. The heat capacity measurements were performed in a custom designed, non-metallic rotatable calorimeter.<sup>5</sup>

This thermodynamic method provides unambiguous evidence for phase transitions in materials. The second technique employed is a tunnel diode oscillator (TDO) circuit.<sup>6</sup> The TDO allows the experimentalist to make contactless measurements on micron-sized samples that are placed in the coil of a self resonant tank circuit. Changes in materials properties will alter the TDO resonance frequency, which in the superconducting state is directly proportional to the penetration depth of the magnetic field. The third method employs torque measurements using a cantilever that carries the sample and acts as one plate of a capacitor. The measured signal is a voltage proportional to the torque, which, in turn, is directly proportional to the magnetization of the specimen. This method can be used to detect changes to better than 1 ppm.

## Results and Discussion

Figure 1 shows heat capacity as a function of field for the field orientation parallel to the planes. The blue curve taken at 425 mK shows a jump at the critical field of 11.1 T. The red curve taken at 250 mK clearly displays a second phase transition at 10.5 T just below the critical field of 11.3 T.<sup>7,8</sup> This additional thermodynamic transition is only present for temperatures below 350 mK and within  $\pm 10^\circ$  of the plane parallel orientation. The lower of the two transitions is the

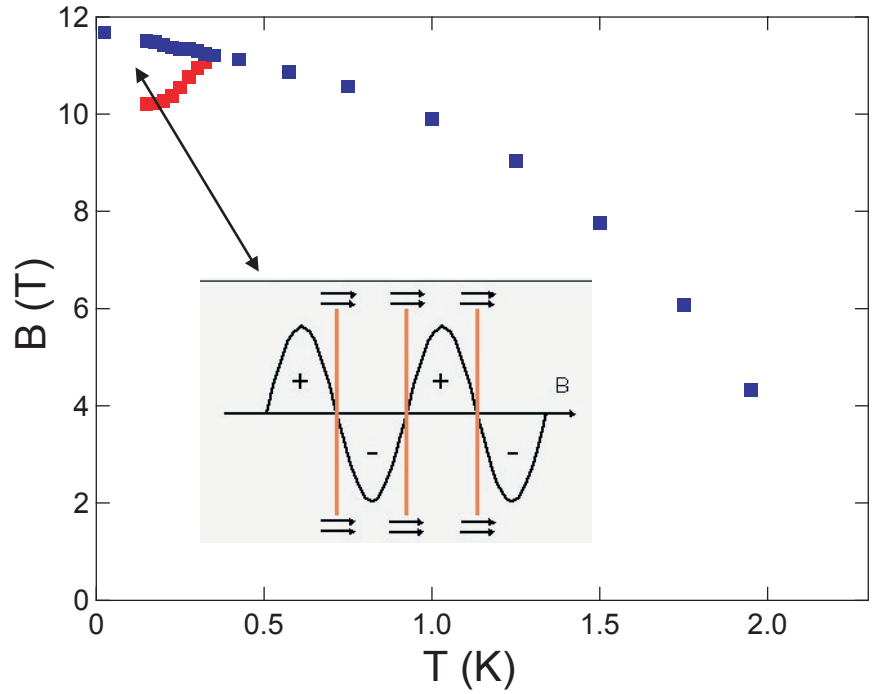


**Figure 1.** Heat capacity showing the additional FFLO phase transition below the critical field at 250 mK (arrow).

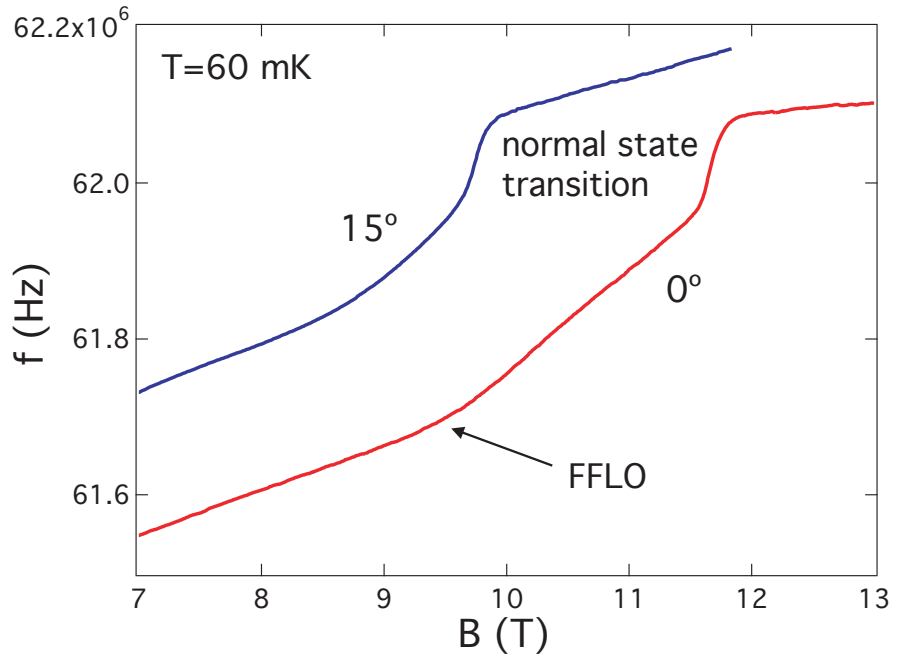
theoretically predicted uniform superconducting-to-non-uniform superconducting (FFLO) state transition. At angles higher than approximately  $10^\circ$ , the orbital effect dominates the paramagnetic effect and FFLO superconductivity cannot be established. This measurement constitutes the first thermodynamic evidence of the non-uniform FFLO state.

In Figure 2, we show the B-T phase diagram as obtained from heat capacity for the plane parallel orientation. The blue squares denote the critical field separating the normal metallic state from the superconducting one. The wedge at low temperatures and high magnetic fields is the FFLO phase, with the red squares separating uniform from non-uniform superconductivity.

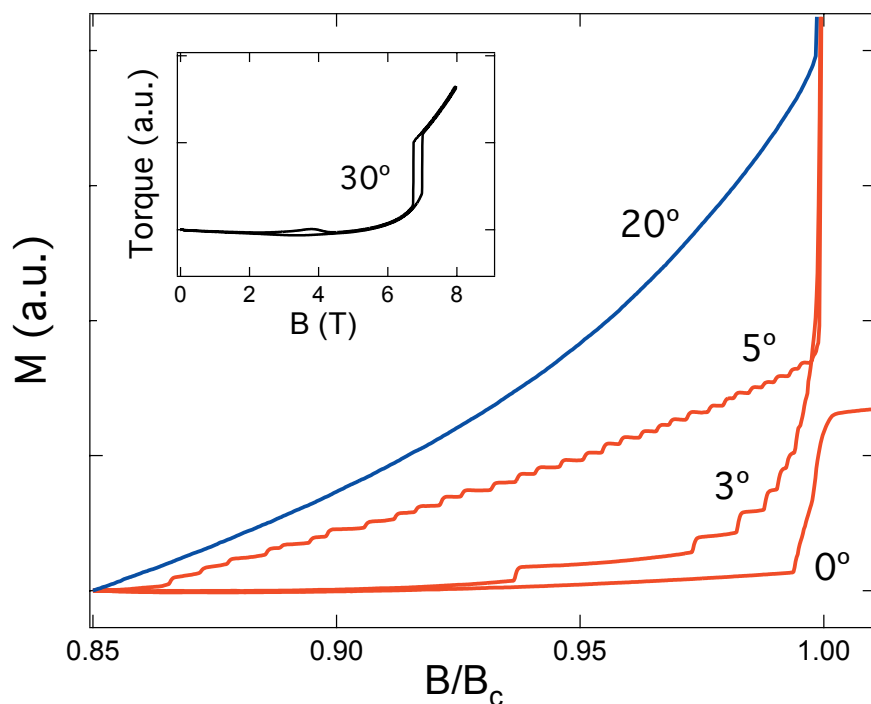
Figure 3 shows our TDO measurements as function of applied field at approximately 60 mK. The red curve is taken with the field oriented parallel to the conducting planes. The jump at 11.7 T is the normal state phase transition, while the additional kink at 9.5 T denotes the FFLO phase transition.<sup>9</sup> The blue curve displays data taken at  $15^\circ$  and only the critical field at 9.8 T is seen, behavior identical to that observed in our heat capacity measurements. The kink in the TDO data matches the appearance of the second phase transition seen in heat capacity in both angle *and* temperature. This provides convincing evidence that the



**Figure 2.** Critical field (blue) and FFLO transition line (red). Inset shows schematically non-uniform FFLO order parameter with magnetic walls. Hysteresis in the heat capacity measurements and the kink and jump nature of transitions in the TDO data confirm that the lower field transition is second order and the higher transition is first order.



**Figure 3.** Penetration depth displaying a kink (arrow) in the plane parallel orientation and identified as the FFLO signature.



**Figure 4.** Magnetization displaying Landau level quantization steps close to the plane parallel field orientation. Inset shows cantilever signal for full field cycle outside FFLO regime. Hysteresis is seen in the field sweeps confirming the first order nature of these transitions.

FFLO transition can be seen in penetration depth studies, too, and will allow us to perform high pressure studies of the FFLO state in the near future.

As discussed in the introduction, the coexistence of orbital and paramagnetic effects can promote higher Landau level states of the order parameter. We show in Figure 4 magnetization as function of normalized magnetic field at  $T = 25$  mK. The divergence at  $B/B_c = 1.00$  is the critical field. At angles slightly out of the planes ( $3^\circ$  and  $5^\circ$ ), a cascade of steps appears, which we identify as transitions into sub-phases with higher Landau quantum numbers.<sup>8</sup> The exact plane parallel orientation cannot support higher Landau states, as the orbital effects die out. The blue curve at  $20^\circ$  represents the uniform superconducting state. The phase space in which these steps are observed exactly matches the FFLO wedge defined by our heat capacity and penetration depth studies.

In this new superconducting state magnetism works to prop up or even enhance superconductivity. These two phenomena were mutually exclusive within the BCS theory and only recently evidence started emerging for their constructive coexistence. We hope that our results will help to shed light on the unique coexistence of magnetism and superconductivity in this and other systems.

## Acknowledgements

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